

**REMARKS**

In the Office Action: claim 3 was allowed; claims 4 and 6-7 were rejected under 35 USC § 112, first and second paragraphs on various grounds; claim 4 was rejected under 35 USC § 102(b) over JP 02-187734A ("Sekisui"); claims 4, 6 and 7 were rejected under 35 USC § 102(b) over J. Phys. Chem. 1993, 97, pp. 9385-9389 ("Puccetti"); and claims 4, 6 and 7 were rejected under 35 USC § 102(e) or 35 USC § 103(a) over USPN 5,912,257 ("Prasad").

In response to the Office Action, claims 4 and 6-7 have been cancelled and represented as claims 16-18, without prejudice. Support for the amendments may be found throughout the application as filed, for example in the original claims and as set forth in previous amendments. No new matter is added.

**Conditional Request for Telephonic Interview**

A telephonic interview is requested prior to the issuance of a subsequent Office Action should any issues remain after entry of this response that would delay allowance of the claims. The undersigned may be reached at (858) 228-7829.

A telephonic interview had initially been scheduled in January 2006, but was canceled due to a PTO scheduling conflict. Attempts at rescheduling were unsuccessful, and the Examiner stated that he wished to communicate in writing regarding some previously uncited art. The current Office Action resulted.

**The Rejections Under 35 USC § 112**

Claims 4 and 6-7 were rejected under 35 USC § 112, first and second paragraphs, on the grounds that certain terms allegedly lacked written description in the application and allegedly rendered the claims indefinite. These rejections are traversed or otherwise addressed below.

**Written Description**

MPEP 2163 (III)(A) sets forth the procedural standard for determining the adequacy of the description of a claimed invention during prosecution:

A description as filed is presumed to be adequate, unless or until sufficient evidence or reasoning to the contrary has been presented by the examiner to rebut the presumption. See, e.g., *In re Marzocchi*, 439 F.2d 220, 224, 169 USPQ 367, 370 (CCPA 1971). The examiner, therefore, must have a reasonable basis to challenge the adequacy of the written description. The examiner has the initial burden of presenting by a preponderance of evidence why a person skilled in the art would not recognize in an applicant's disclosure a description of the invention defined by the claims. *Wertheim*, 541 F.2d at 263, 191 USPQ at 97.

The written description standard simply requires that one of skill can recognize the identity of the claimed subject matter in the disclosure. No particular form of disclosure is required.

[T]he language of the specification, to the extent possible, must describe the claimed invention so that one skilled in the art can recognize what is claimed. ... The disclosure must allow one skilled in the art to visualize or recognize the identity of the subject matter purportedly described. *Enzo Biochem, Inc. v. Gen-Probe, Inc.*, 296 F.3d 1316, 1328, 1329 (Fed. Cir. 2002), internal citations omitted.

Thus, the claimed invention is presumptively described, and the Office has the burden of establishing by a preponderance of evidence why one of skill can not recognize in the application what is claimed.

**Definiteness and Distinct Claiming Under 35 USC § 112, Second Paragraph**

"The requirement to 'distinctly' claim means that the claim must have a meaning discernible to one of ordinary skill in the art when construed according to correct principles. Only when a claim remains insolubly ambiguous without a discernible meaning after all

reasonable attempts at construction must a court declare it indefinite." *Metabolite Labs., Inc. v. Lab. Corp. of Am. Holdings*, 370 F.3d 1354, 1366, 71 USPQ2d 1081, 1089 (Fed. Cir. 2004).

As set forth in MPEP 2173.02, a claim term is definite if its meaning is discernible. *Bancorp Services, L.L.C. v. Hartford Life Ins. Co.*, 359 F.3d 1367, 1372, 69 USPQ2d 1996, 1999-2000 (Fed. Cir. 2004).

Only if a claim term is insolubly ambiguous after all reasonable efforts at construction can it be declared indefinite.

**Consistent use of commas and semicolons.** Applicants were requested to revise the claims for consistent use of commas and semicolons. Applicants have accordingly restructured the claims for clarity and have consistently used commas and semicolons in the revised claims.

**“Derivatives thereof” and “derived essentially from.”** Claims 4 and 6-7 were rejected as allegedly lacking written description and as allegedly indefinite for the use of these terms in the definition of Ra1, Ra2, and Ra3, as well as in the definitions of Re-Rm and Rg1-3. These rejections are traversed. The claims as examined meet the statutory standards, and were sufficiently described and sufficiently clear for a search to be performed. Nevertheless, the claims have been revised in order to advance prosecution, and no longer recite these terms.

Applicants extensively discuss substituting the chromophores in order to alter their properties throughout the application. For example, at page 18, improvement of lipophilicity is discussed by introducing dibutyl or dibutylphenyl groups, while hydrochloride acid adducts of bis-lysyl esters of the chromophores are described as hydrophilic, and fluorescent in solution. Substitution of the chromophores with surface specific functionalities is also discussed. Amino-acid substituted chromophores are also discussed at page 36, and substituents are also described for the functional groups at pages 22-27, and throughout the examples.

It can be recognized by one of skill that the applicants describe substitution of these positions with functional groups to alter chromophore properties. The definitions of these substituents in the application list a variety of functional groups that can be introduced.

The claims have therefore been revised to recite that the substituents at these positions, in addition to H or alkyl groups, can be a functional group obtained by reaction with those species described in the application as filed: an amino acid, a polypeptide, adenine, guanine, tyrosine, cytosine, uracil, biotin, ferrocene, ruthenocene, cyanuric chloride, or methacryloyl chloride. The claims no longer recite derivatives of these described groups.

The revised claims meet the written description and definiteness standards, and accurately express the invention set forth in the application. As the terms objected to have been removed, withdrawal of these rejections are respectfully requested.

**“Polymerizable functionalities.”** This term was said to lack description and examples of species of  $R_{e-m}$ , and to therefore be indefinite. Applicants traverse these rejections. The definition of  $R_{e-m}$  at page 24 lines 23-24 refers to “NOTE 2A” for the definition of polymerizable functionalities. Polymerizable functionalities are extensively described and exemplified in NOTE 2A at page 23 line 18 through page 24 line 14. Polymerizable functionalities were said to include vinyl, allyl, 4-styryl, acroyl, methacroyl, acrylonitrile, isocyanate, isothiocyanate, epoxides, and strained ring olefins. In order to expedite prosecution, the revised claims recite these specific embodiments in place of this term. Applicants reserve the right to pursue claims using the generic language.

#### **The Cited Art Rejections**

Claim 4 was rejected under 35 USC §102(b) over JP 02-187734A (“Sekisui”); claims 4, 6 and 7 were rejected under 35 USC §102(b) over Puccetti (J. Phys. Chem. 1993, 97, pp. 9385-9389); and claims 4, 6 and 7 were rejected under 35 USC §102(e) or 35 USC §103(a) over USPN 5,912,257 (“Prasad”). These rejections are traversed. The art cited in the Office Action does not teach or suggest the invention as claimed. In fact, the cited art itself teaches distinctions between the nonlinear optical phenomena forming the basis for the alleged inherency and the

multiphoton absorption techniques recited in the claims. Furthermore, the Office Action does not meet the burden in establishing inherency over these publications.

As set forth below, Sekisui and Puccetti discuss only nonlinear optical phenomena, and Prasad discusses compounds not falling within the claim scope. None of these publications teach or suggest multiphoton absorption by any compound within the claims. Thus, the cited art lacks all the claim elements, and cannot support either an anticipation or an obviousness rejection.

**As the cited art describes, optical harmonic generation and nonlinear optical processes are unrelated to multiphoton absorption.** Prasad, cited in the Office Action, teaches that nonlinear optical phenomena such as harmonic generation are distinct from two- or multiphoton absorption methods (Prasad, col. 1 lines 36-45, emphasis added):

Frequency upconversion lasing is an important area of research and has become more interesting and promising in recent years. Compared to other coherent frequency upconversion techniques, such as optical harmonic generation or sum frequency mixing based on second- or third-order nonlinear optical processes, the major advantages of upconversion lasing techniques are: i) elimination of phase-matching requirements, ii) feasibility of using semiconductor lasers as pump sources, and iii) capability of adopting waveguide and fiber configurations. To date, two major technical approaches have been used to achieve frequency upconversion lasing: one is based on direct two-photon (or multi-photon) excitation of a gain medium (two-photon pumped); the other is based on sequential stepwise multi-photon excitation (stepwise multi-photon pumped).

The cited art clearly teaches that two or multi-photon excitation is an alternative to optical harmonic generation or nonlinear optical processes method for upconverting frequencies. Discussions of nonlinear optical processes in the cited art thus have no bearing on whether the materials discussed possess two-photon absorption capabilities.

**Second harmonic generation results from an electrical interaction, not from photon absorption.** Second harmonic generation results from the interaction of the electrical field of an

intense incident beam of electromagnetic radiation with the electrons in a material, not from absorption of multiple photons. The outermost electrons in the material are induced by the beam to form a small electric dipole pointing in the direction of the impressed electronic field, oscillating with that field.

As can be seen in the derivation of Equations 4-8 at page 5 of Appendix A, the induced charge polarization in a material contains a first (major) component of the charge polarization which is proportional to the angular frequency of the incident light (see Equation 8), while the second component is proportional to twice the angular frequency.

Thus, a rapidly oscillating incident beam induces two types of oscillating charge polarizations in a material: one with the frequency of the incident light, and one of twice that frequency. These oscillating dipoles then interact with incident photons and absorb and reradiate them. Some of the light that interacts with a dipole oscillating electronically at twice the frequency of the incident light is reradiated at that frequency. This is what leads to second harmonic generation, not the absorption of two photons.

**Sekisui.** A very brief partial translation was cited in the Office Action as allegedly supporting the inherency of multiphoton absorption in Sekisui. The partial translation purportedly described stimulation of the material with a pulsed laser (“pulsed” written in column 12), having an apparent wavelength of 1.064  $\mu\text{m}$ . The phrase “3x higher than urea” is marked in the margin of column 12, but is not explained.

Sekisui also apparently describes “SHG” (see col. 12), presumably second harmonic generation, and mentions 532 nm, which is exactly the wavelength expected from second harmonic generation, a nonlinear optical process. As set forth above, nonlinear optical

phenomena, including second harmonic generation, are unrelated to multiphoton absorption, and Sekisui's discussion of second harmonic generation cannot support an inherency rejection of the multiphoton absorption-based methods claimed here.

**Puccetti.** Similarly, Puccetti discusses only nonlinear optical processes, and does not teach or suggest multiphoton absorption by the materials used.

Regarding Puccetti, the Office Action stated that “[t]he relaxation of the excited state to emit 2x or 3x the energy of the incident radiation / at least  $\frac{1}{2}$  the wavelength is evidence that at least some simultaneous multi-photon absorption occurs in the process as inherent to the production of the second harmonic generation resulting from the pulsed laser irradiation of the same compounds.” Office Action, page 6.

This statement is not accurate. The emission of light with half the wavelength in nonlinear optical processes has no relationship whatsoever to how many photons a material absorbed. Because these phenomena are unrelated, Puccetti cannot support an inherency rejection of the claimed multiphoton absorption-based methods.

Furthermore, Puccetti teaches that his compounds are *transparent* at the wavelengths employed. See page 9388, right column (the harmonic wavelength ... is located within the transparency range of the molecules). Thus the molecules used by Puccetti do not absorb at  $\frac{1}{2}$  or  $\frac{1}{3}$  the wavelength of the incident light.

**The Office Action does not meet the burden for establishing inherency.** Anticipation requires that all claim elements be present in a single reference. Anticipation by inherency requires that each claim element not explicitly taught must necessarily be present in that

reference. The Patent Office bears the burden of showing that an allegedly inherent property is necessarily present in the cited reference (MPEP 2112):

"In relying upon the theory of inherency, the examiner must provide a basis in fact and/or technical reasoning to reasonably support the determination that the allegedly inherent characteristic *necessarily* flows from the teachings of the applied prior art." Ex parte Levy, 17 USPQ2d 1461, 1464 (Bd. Pat. App. & Inter. 1990) (overturning inherency rejection unsupported by factual basis or cogent scientific reasoning; citations omitted, emphasis in original).

"To establish inherency, the extrinsic evidence must make clear that the missing descriptive matter is necessarily present in the thing described in the reference, and that it would be so recognized by persons of ordinary skill. Inherency, however, may not be established by probabilities or possibilities. The mere fact that a certain thing may result from a given set of circumstances is not sufficient." In re Robertson, 169 F.3d 743, 745, 49 USPQ2d 1949, 1950-51 (Fed. Cir. 1999) (citations omitted).

As recognized by those of skill, including Prasad, nonlinear optical phenomena bear no relation to multiphoton absorption. Thus Sekisui and Puccetti, cited solely for discussing nonlinear optical phenomena, do not and cannot necessarily teach or suggest multiphoton absorption. Puccetti in fact teaches that his molecules are transparent at 1/2 or 1/3 the wavelength of the incident light, and thus exhibit no absorption at these wavelengths. The burden for establishing an inherency rejection of the claimed methods, which utilize multiphoton absorption, has not been met for either publication.

**Prasad.** Prasad's cited compound has an electron accepting group at the position requiring an electron donating group in the claimed methods. See the definition of W at col. 17 lines 21-34, as noted in the Office Action at page 8. Prasad is discussing donor-pi-acceptor molecules (D-Π-A), rather than the D-Π-D (donor-pi-donor) compounds within the claims. See also column 10 lines 45-59. The Office Action asserts that Prasad reads on the claimed groups in



Re-m; however, that position in the claims corresponds to the donor groups –Da(RaRb) or –  
Db(RcRd), not Re-m.

Thus Prasad lacks the claim element of an electron donating group at the position required by the claims. Prasad therefore cannot support an anticipation or obviousness rejection.

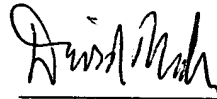
**Withdrawal of the cited art rejections is requested.** Sekisui and Puccetti describe a method unrelated to multiphoton absorption, and cannot inherently anticipate the claims. Prasad describes compounds outside the claim scope. None of these publications teach or suggest the claimed invention. Withdrawal of the rejections is respectfully requested.

CONCLUSION

As the claims are believed in order for allowance, a notice to that effect is respectfully requested. Should the Examiner disagree, a telephonic interview is conditionally requested prior to the issuance of a subsequent Office Action so that any remaining issues can be discussed. The undersigned may be reached at (858) 228-7829.

The Commissioner is hereby authorized to charge any underpayment or credit any overpayment associated with this communication to Deposit Account No. 22242 as necessary for this Response.

Respectfully submitted,



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David W. Maher  
Reg. No. 40,077

## **APPENDIX A**

# MODULE 6-11

# NONLINEAR MATERIALS

General Comments of Leno Pedroitti

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(1) Nonlinear optical materials are materials in which the intensity of light input, including its frequency, is not related to the intensity of light output by a simple proportionality constant. Because of this nonlinear behavior, an intense light beam propagating through a *nonlinear* optical material will produce new effects that can't be seen with weak light beams. For example, an intense light beam propagating through a nonlinear material can generate, in addition, harmonics or overtones of the original light frequency. This means that the red beam from a ruby laser can create an ultraviolet beam as it passes through the nonlinear optical material, while itself still propagating as a red beam.

(2) The interaction of intense light beams with nonlinear materials has opened a large field of potential applications in optical communication systems, where many well-known radio-frequency techniques such as mixing, heterodyning, and modulation now can be transferred to the domain of optical frequencies. For these reasons, nonlinear materials play an ever-increasing role in laser applications.

(3) This module will familiarize you with the fundamental physical ideas that underlie some of the more commonly encountered nonlinear phenomena and how these phenomena depend on the properties of the nonlinear materials. The experiment at the end of this module will give you practical experience in using a nonlinear material to generate the visible second harmonic of the invisible 1.06- $\mu\text{m}$  Nd:YAG laser wavelength.

(4) For a better understanding of the ideas in this module, you should have knowledge of algebra and some trigonometry. Before beginning the experiment at the end of this module, be

sure to review laser safety.



## OBJECTIVES

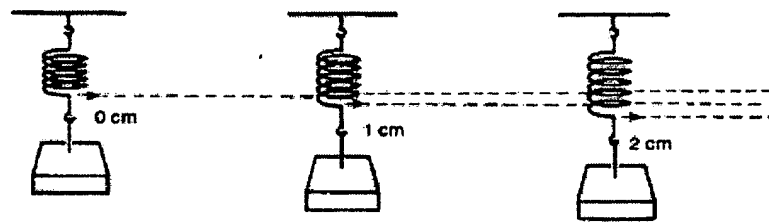
(5) When you complete this module, you should be able to do the following:

- Describe the nonlinear behavior of a system by describing its linear and nonlinear behavior with appropriate equations.
- Explain second harmonic generation of light with the aid of the induced charge polarization.
- Discuss in general terms the need for phase matching in second harmonic light generation.
- Describe two experimental techniques used for phase matching.
- List the various materials and experimental parameters that determine the efficiency of second harmonic generation through the use of the correct mathematical equations.
- Differentiate between passive and active nonlinear materials.
- List and rate the importance of some of the more common nonlinear materials.
- Define the field-dependent nonlinear index and its implications.
- Produce the second harmonic ( $0.53\ \mu\text{m}$ ) of the  $1.06\text{-}\mu\text{m}$  radiation from a Nd:YAG laser using a crystal of KDP or ADP.
- Knowing the type of SHG crystal, its size and operating parameters, estimate the theoretical conversion efficiency.

## DISCUSSION

(6) Our objective is to develop a familiarity with the behavior and application of nonlinear materials. We'll do this by first describing the behavior of a linear system or material. Next, we'll look at its nonlinear behavior and discuss the internal properties of the material that make it behave in a nonlinear fashion. Finally, we'll describe in a very elementary fashion the fundamental physical idea behind one of the more common nonlinear effects and how this effect depends on the properties of the nonlinear material. We conclude this section with a brief discussion of the more common nonlinear materials.

(7) Let's start then with a brief discussion of linear materials and linear effects. It is a fact that our everyday experiences tell us that we live in an essentially linear world. When *you* first encounter the mathematical expressions of elementary physical laws, you may be tempted to conclude that nature is linear. For example, you learn that gas pressure in a container will double if the temperature of the gas is doubled; or that when a given force on a spring produces a stretching of the spring of one centimeter, then doubling the force will produce a stretching of exactly two centimeters. The example of the stretching spring is shown schematically in Figure 1.



**Fig. 1**  
Linear displacement of a spring

(8) From these and other examples you might conclude that the response to small disturbances in all physical systems follows a linear law. Doubling the cause doubles the effect, tripling the cause triples the effect, and so on.

(9) Consider once more a linear spring. The mathematical expression relating the force  $F$  on the spring to the stretching of the spring in the  $x$ -direction is

$$F = kx \quad \text{Equation 1}$$

where the proportionality constant  $k$  is the spring constant.

(10) In practice, as long as the stretching of the spring is small compared to the total length of the spring, the force can be calculated with Equation 1. However, when the distance through which the spring is stretched exceeds a certain limit, the magnitude of the force  $F$  no longer is related to the distance  $x$  in a simple linear manner. Then it's more accurately expressed by the nonlinear relation.

$$F = kx + k'x^2 + k''x^3 + k'''x^4 + \dots \quad \text{Equation 2}$$

where  $k'$ ,  $k''$ ,  $k'''$  are higher order constants and are much smaller than the constant  $k$ .

(11) The magnitude of the force as a function of the distance the spring is stretched is shown in Figure 2, where the linear as well as the nonlinear behavior of the spring is illustrated.

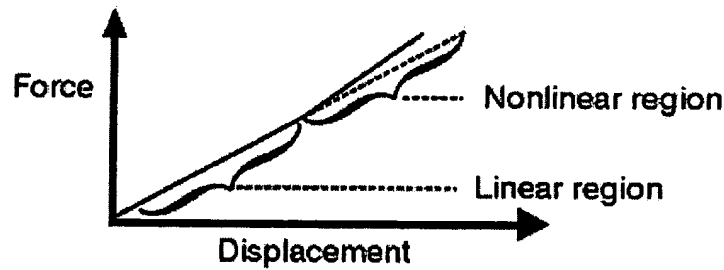


Fig. 2  
Linear and nonlinear behavior at a stretched spring

(12) Now we've stated what we mean by "linear" and "nonlinear" behavior of a system (the spring). Let's next discuss the nonlinear behavior of certain materials when intense light beams propagate through them. These light beams are of course electromagnetic waves that are made up of an electric field and a magnetic field. Both fields are locked in step at right angles to each other and oscillate together at the frequency of the light, as you have already learned.

(13) Consider what happens when such an oscillating field interacts with an atom, which you can think of as a positively charged nucleus surrounded by shells of electrons. The electrons in the outermost shell are more loosely bound, and their radial displacements are governed by the same force equation that describes the stretching of a spring. These outermost electrons can redistribute themselves in step with the electric field. In this way the atom becomes charge-polarized. This means that positive and negative parts separate slightly to form a small electric dipole with a so-called *dipole moment* pointing in the direction of the impressed field. The dipole moment is the magnitude of the displaced charge times the distance between the positive nucleus and the center of the displaced electron shell. (Note that when not displaced, the center of a spherical electron shell is, of course, at the same location as the positive nucleus.)

(14) The number of electric dipoles per unit volume multiplied by the dipole moment of one atom gives the induced *macroscopic charge polarization* of the material. (Don't confuse this charge polarization induced by the electric field of the light beam with the polarization of a light beam with a polarizing filter.)

(15) In any given material, the magnitude of the induced charge polarization depends on the magnitude of the applied electric field  $E$ . Specifically, we can express the charge polarization  $P$  in a series of powers of  $E$  and write

$$P = aE + dE^2 + d'E^3 + \dots \quad \text{Equation 3}$$

where:  $a$  = Polarizability coefficient of the material

$d, d'$ , etc. = Higher-order nonlinear optical coefficients that are much less than  $a$

The similarity between Equation 3 and Equation 2 should be clear.

(16) The electrons in the outermost shell not only are subject to the pulling of the applied

external field, but also are subjected to the pull of the internal field due to the positive nucleus. The magnitude of this internal electric field is enormously large, somewhere in the neighborhood of  $10^9$  volts/cm. For comparison, the electric field of sunlight at the surface of the earth is only about 10 volts/cm. It turns out that, when the applied electric field is many orders of magnitude less than the atom's internal electric field—which is always the case for ordinary light sources—the linear approximation  $P = aE$  of Equation 3 is very accurate.

(17) With the development of the laser in 1960 we were able to build light sources capable of producing optical electric fields of  $10^6$  to  $10^7$  volts/cm. When some optical materials are irradiated by such intense fields, the contribution to the charge polarization by the higher-order terms in Equation 3 no longer can be neglected.

(18) Let's see what happens when we use only the first two terms in Equation 3. For simplicity, let's also assume that the electric field points in the x-direction. Then the total induced charge polarization in the x-direction is

$$P_x = aE_x + dE_x^2 \quad \text{Equation 4}$$

(19) For plane polarized light of frequency  $f$  and amplitude  $E_{xo}$ , the electric field at some given point in an optical material is described by

$$E_x = E_{xo} \cos(2\pi ft) = E_{xo} \cos(\omega t) \quad \text{Equation 5}$$

where  $\omega \equiv 2\pi f \equiv$  the "angular frequency" in rad/sec

and the induced charge polarization is

$$P_x = aE_{xo} \cos(\omega t) + dE_{xo}^2 \cos^2(\omega t) \quad \text{Equation 6}$$

Using the trigonometric identity

$$\cos^2\theta = \frac{1 + \cos 2\theta}{2} \quad \text{Equation 7}$$

Equation 6 can be written as:

$$P_x = aE_{xo} \cos(\omega t) + \frac{dE_{xo}^2 \cos 2(\omega t)}{2} + \frac{dE_{xo}^2}{2}$$

or

$$\text{Equation 8}$$

$$P_x = P_x(\omega) + P_x(2\omega) + \text{constant term (DC)}$$



(20) An inspection of the above equation reveals that the first term  $P_x(\omega)$  is a charge polarization of angular frequency  $\omega$ . The second term  $P_x(2\omega)$  is a charge polarization of angular frequency  $2\omega$ , twice the fundamental frequency  $\omega$ . Finally, the third term has no frequency dependence whatsoever. It provides a "DC" charge polarization. The three charge polarization terms are shown in Figure 3. Equation 8 then tells us that a rapidly oscillating electric field in the neighborhood of  $10^{14}$  to  $10^{15}$  Hz (optical frequencies) will induce two types of oscillating charge polarization; one of frequency  $\omega$  and another of frequency  $2\omega$ . The material consists, then, of a collection of oscillating dipoles that absorb and reradiate light waves. The total energy in the beam of light is not significantly altered.

(21) One effect of the induced charge polarization and subsequent reradiation is a decrease in the speed of the light in the medium. The decrease in speed is reflected in the increased value of the index of refraction  $n$ , which is given by the ratio of the speed of light in vacuum  $c$  divided by the speed of light in the medium  $v$ , that is, Equation 9,

$$n = \frac{c}{v}$$

Equation 9

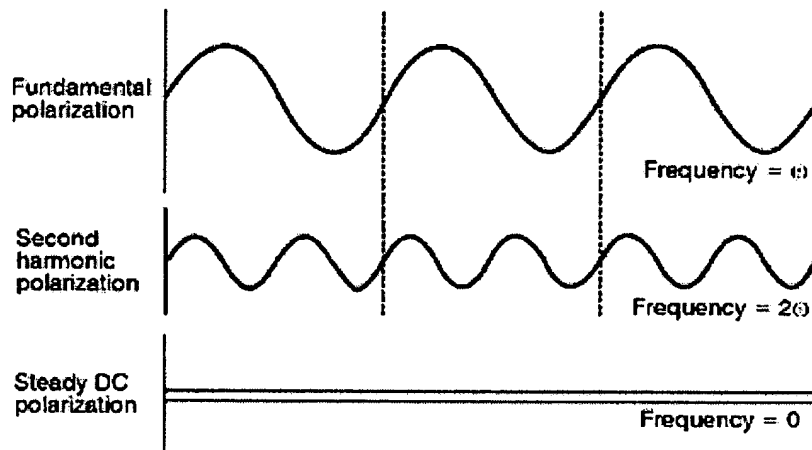


Fig. 3  
Frequency associated with charge-polarization terms

(22) Another effect, predicted by Equation 8, is the reradiation of energy at the frequency  $2\omega$ , twice the frequency of the incident radiation. This doubling of frequency is known as "second harmonic generation" (SHG) or "frequency doubling."

(23) Frequency doubling was observed for the first time in 1961 by Professor Peter Franken and some graduate students at the University of Michigan. They irradiated a quartz crystal with the beam from a ruby laser that operated at 694.3 nm. A very small amount of the light striking the crystal was converted to light with a wavelength of 347.2 nm. This wavelength lies in the ultraviolet region of the spectrum and is of course exactly half the wavelength and twice the frequency of the incident laser light.

(24) The details of this famous experiment are shown in Figure 4. As you might expect, this

experiment initiated a search for materials in which this effect occurs strongly. As a result, the experiment has been duplicated many times with a host of different nonlinear materials.

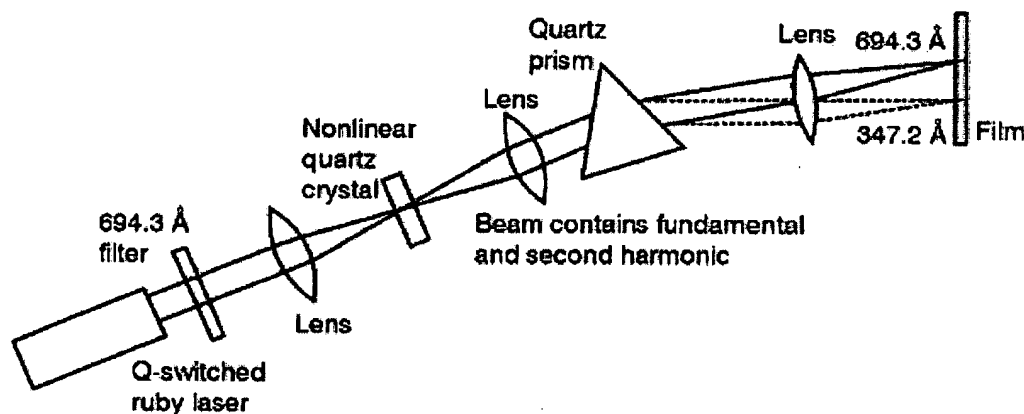


Fig. 4

Experimental arrangement for second harmonic generation

(25) What are the properties of a crystal that are responsible for efficient SHG? To answer this question let's first note that, whatever these properties may be, they manifest themselves in the expansion coefficient  $d$  of the second-order term in Equation 3.

(26) To find out how  $d$  is governed by the internal properties of the crystal, we need to examine the oscillatory motion of the dipoles. This is best done by adding the three curves in Figure 3. The resultant charge polarization is shown in Figure 5, along with the applied electric field.

(27) The curve for the total polarization shows mainly the contributions of the fundamental polarization and steady DC polarization terms. The contribution of the second harmonic term would cause a slight change in the slopes of the rise and fall of the total polarization, but not in the location of the maximum, zero, and minimum points. So its effect has not been shown in the lower curve in Figure 5.

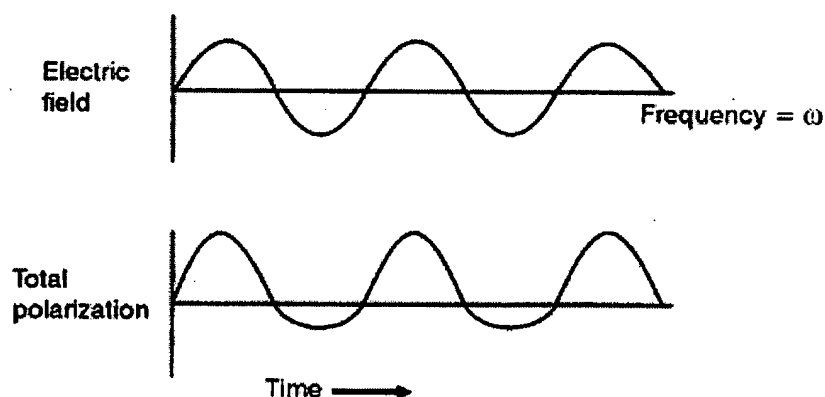


Fig. 5

Impressed electric field and resulting total polarization

(28) It's clear from Figure 5 that the individual dipoles, whose oscillatory motions lead to the charge polarization, do not faithfully reproduce the sinusoidal electric field. Note especially

that the total polarization is asymmetric in the upward direction. This is because the outer electrons of the atoms in a crystal are more easily pushed in one direction than in the opposite direction.

(29) Figure 5 shows that an intense electric field in the up-direction is more effective in polarizing the medium than the same field in the down-direction. Such a situation can occur only in a crystal with no internal symmetry. Only about 10% of the crystals found in nature fall in this category.

(30) The distorted charge polarization wave produced by the intense radiation field travels at the same speed as the incident light wave. This light, which is reradiated by the oscillating dipoles, contains, as we saw above, two discrete frequencies  $\omega$  and  $2\omega$ . Since the refractive index for the light of frequency  $2\omega$  is usually larger than it is for light of frequency  $\omega$ , the speed of the component with frequency  $2\omega$  will be just a bit slower than the component with frequency  $\omega$ , in accordance with Equation 9.

(31) The difference in propagation speed between the fundamental frequency and the second harmonic frequency means that the second harmonic light radiated from one part of the crystal, say  $P(1)$ , will not be in phase with the second harmonic light radiated from another part of the crystal  $P(2)$ . As a result, the two waves from  $P(1)$  and  $P(2)$  will interfere destructively at some point  $P(3)$ . The distance that it takes for a second harmonic charge polarization wave and its radiated light to get  $180^\circ$  out of phase is called the "coherence length." This distance is usually on the order of  $10 \mu\text{m}$ . If the crystal length happens to be equal to an odd multiple of the coherence length, virtually no second harmonic radiation will emerge from the crystal.

(32) This problem of destructive interference can be overcome by making use of the double-refraction birefringence exhibited by certain crystals. It turns out that in such crystals the velocity of light depends on the direction of polarization of the electric field, the propagation direction, and the wavelength.

(33) For example, the speed of light in the nonlinear material potassium dihydrogen phosphate (KDP) for the horizontally and perpendicularly polarized electric field components is shown in Figure 6. As you can see, the ordinary fundamental that we have taken to be the light from a ruby laser at  $694.3 \text{ nm}$ , polarized perpendicular to the plane of the page, travels at exactly the same speed as the extraordinary second harmonic light at  $347.2 \text{ nm}$ , when both light waves propagate in a direction that's inclined  $50^\circ$  to the optic axis of the crystal. The optic axis of a doubly refracting crystal is that direction along which the ordinary and extraordinary waves of the same frequency propagate at the same speed.